

Magneto-electric and Magneto-dielectric Effects in REMn_2O_5

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[*Nature* 429, 392 (2004)]
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[*Phys. Rev. Lett.* (2004) in print]
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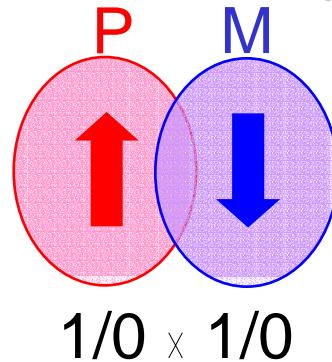
I. MULTIFERROIC

Coexistence of two or more properties of
(anti)ferroelectricity, (anti)ferromagnetism and (anti)ferroelasticity

Applications

- Multiple state memory
- Write to E / Read from M
- Magnetically switchable optical device

Ferroelectromagnets



- Additional degree of freedom
- Coupling between P&M

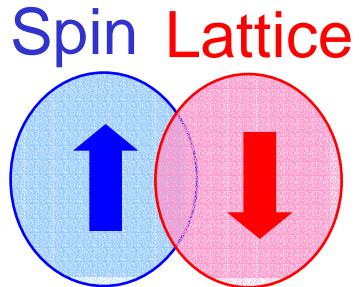
♣ A few candidate materials

We need more materials for better understandings!!!

Known materials

- BiMnO_3
- BiFeO_3
- $\text{Ni}_3\text{B}_7\text{O}_{13}\text{I}$
- Hexagonal $R\text{MnO}_3$
($R=\text{Ho-Lu}$)

“Spin-Lattice coupling”



Order parameter: M
Driving force: H
Linear coefficient: χ

“1. Magneto-electric effect”

Induction of magnetization by electric field (ME_E effect)
Induction of polarization by magnetic field (ME_H effect)

ME_H effect

$$P_k(\vec{E}, \vec{H}; T) = -\frac{\partial g}{\partial E_k} = \dots + P_k^s + \varepsilon_0 \varepsilon_{ik} E_i + \alpha_{ki} H_i + \frac{1}{2} \beta_{kij} H_i H_j + \gamma_{ijk} H_i E_j + \dots$$

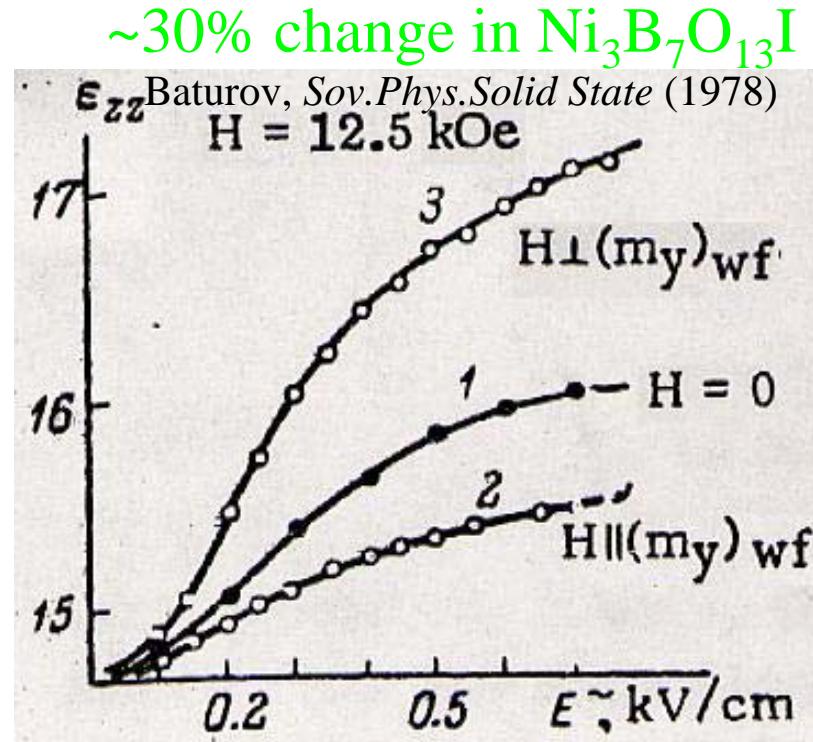
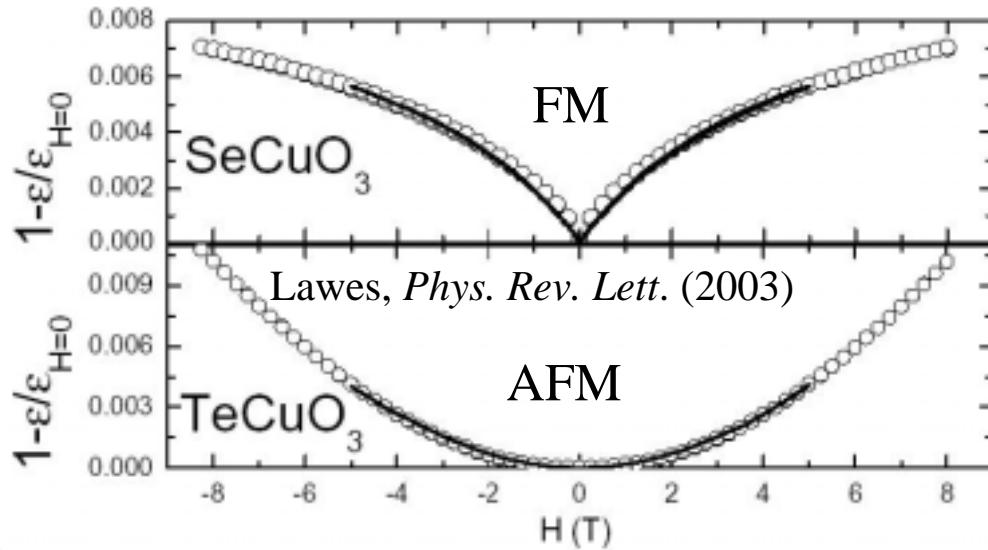
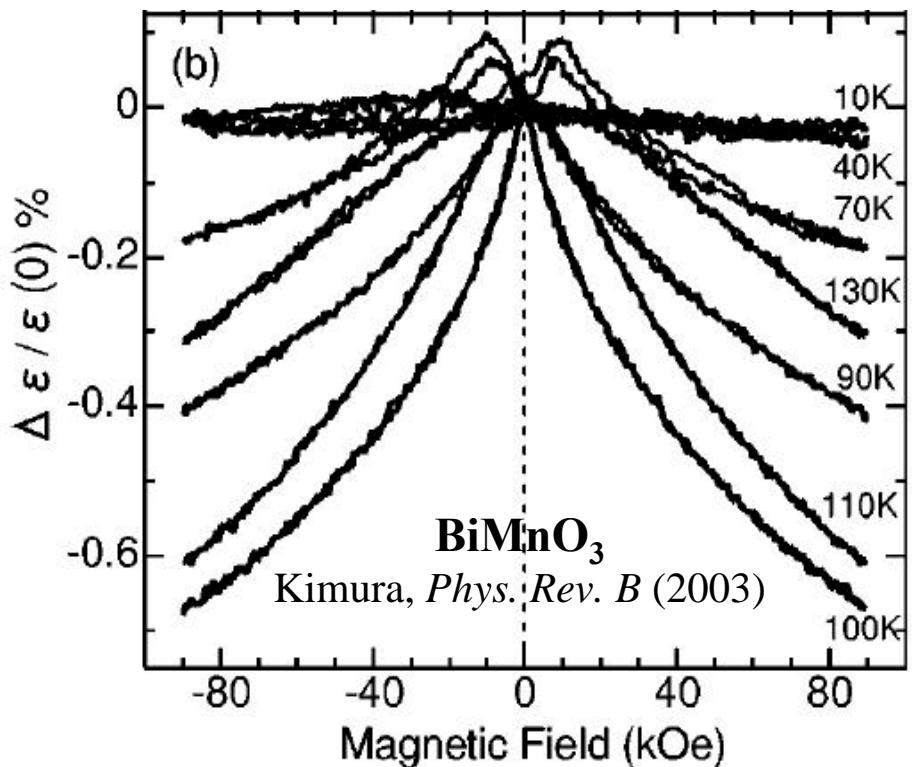
Total polarization Spontaneous P linear ME_H effect bilinear ME_H effect

“2. Magneto-dielectric effect”

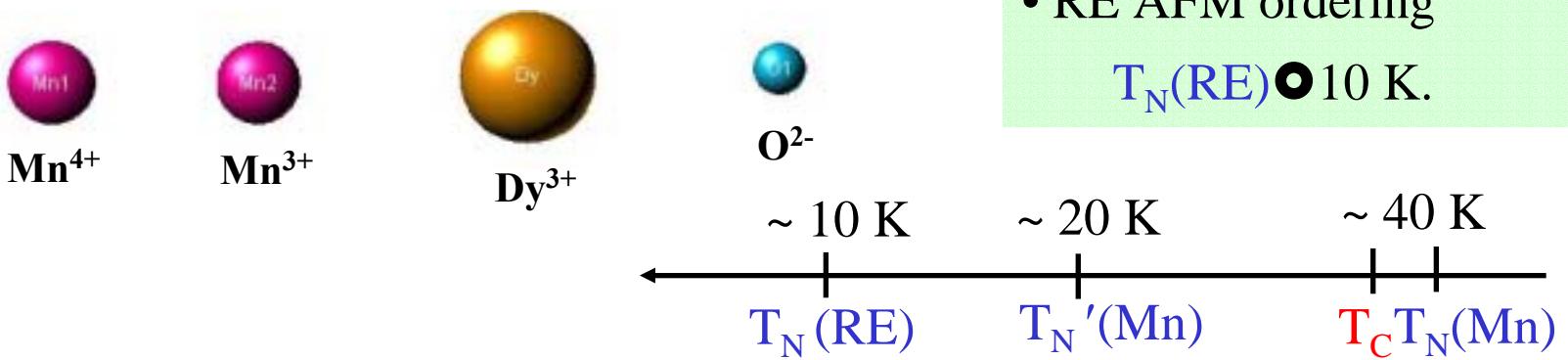
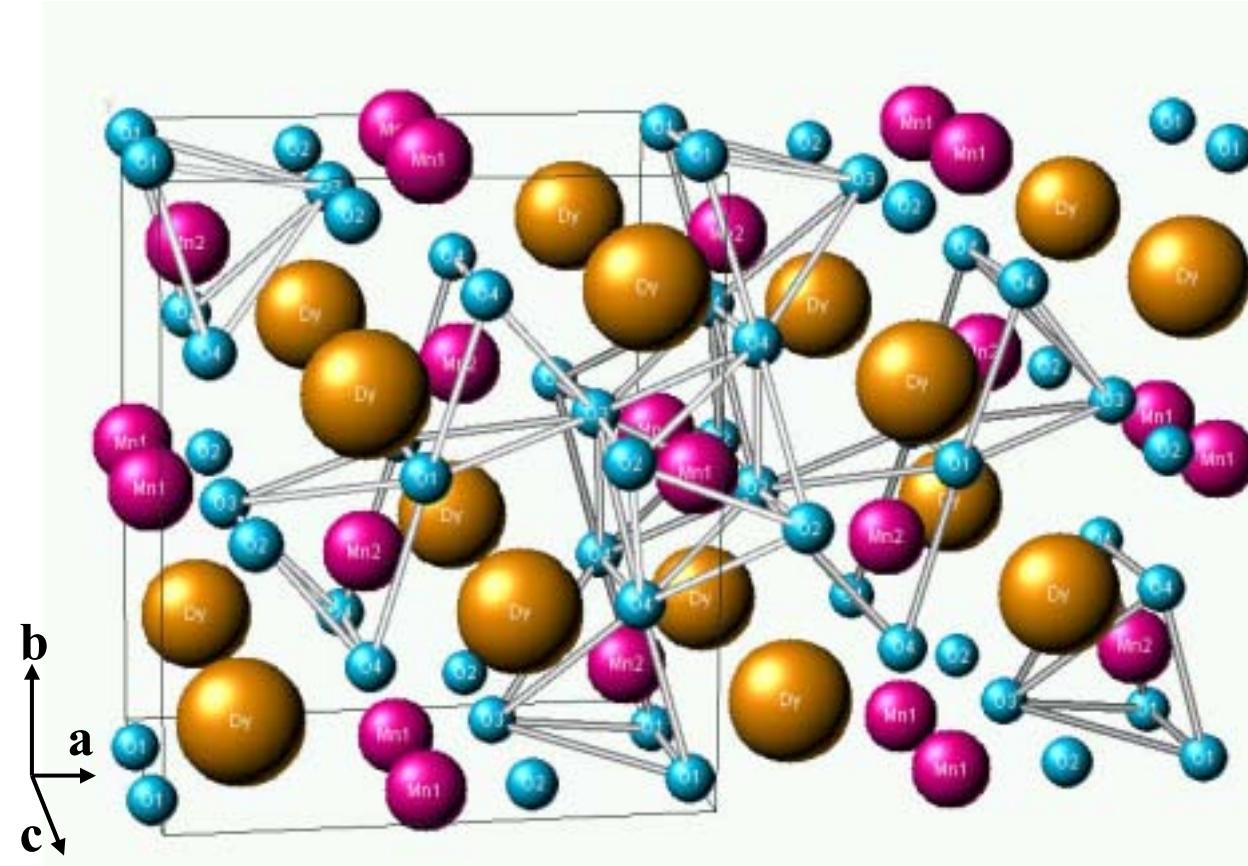
- ♣ Dielectric constant change by H
- ♣ Magnetically tunable capacitor

Ginzberg-Landau theory

$$\Omega \varepsilon \sim \eta_0 M^2$$



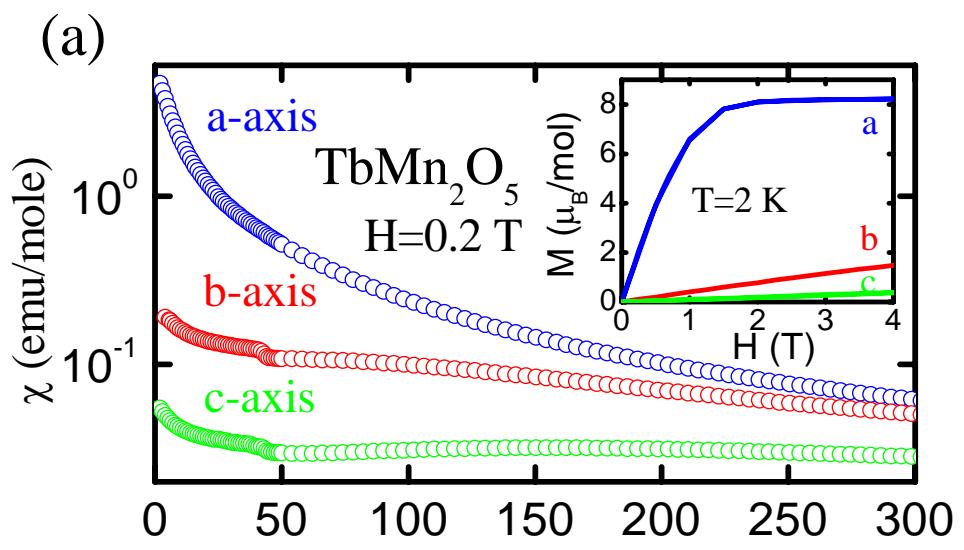
What's known about REMn_2O_5 ?



- Orthorhombic structure (space group: $Pbam$)
- Mn^{4+} : at the center of oxygen octahedron
- Mn^{3+} : near the base center of a square pyramid
- FE along b , $\text{T}_C \sim 35-40 \text{ K}$.
- AFM, $\text{T}_N(\text{Mn}) \sim 40 \text{ K}$
- Mn spin reorientation $\text{T}_N' \sim 20-25 \text{ K}$
- RE AFM ordering $\text{T}_N(\text{RE}) \bullet 10 \text{ K}$.

III. TbMn₂O₅

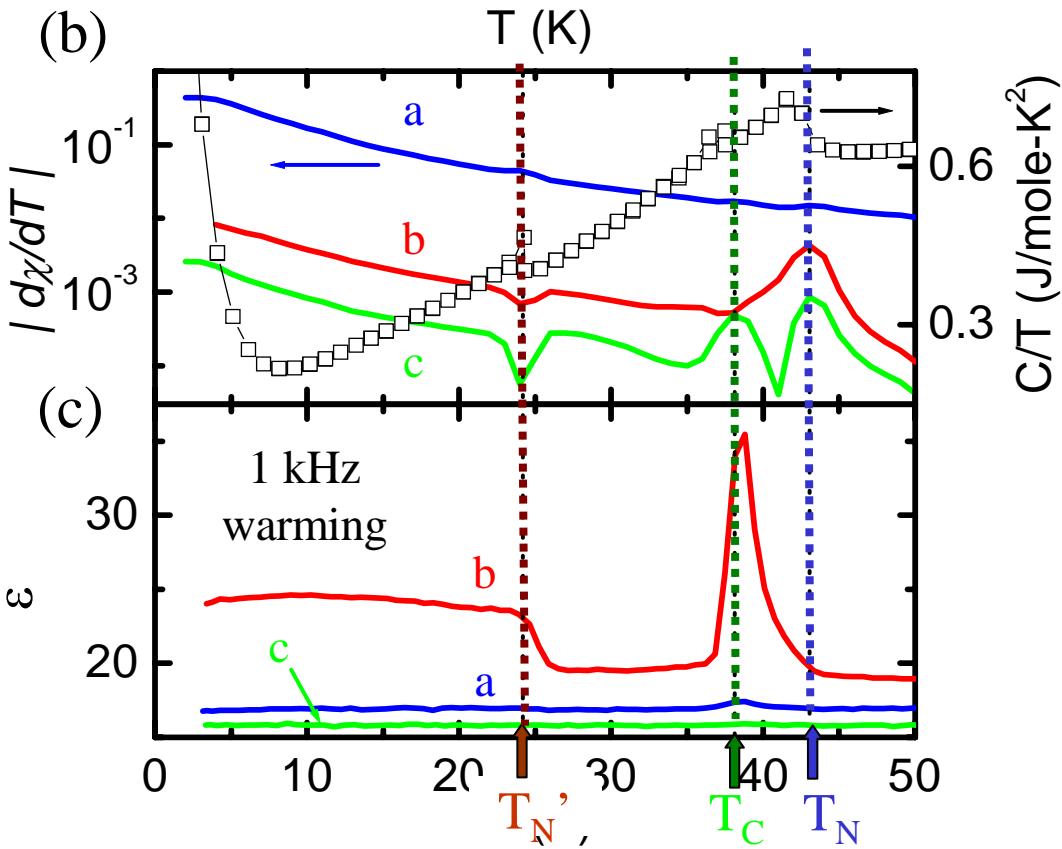
(a) •large magnetic anisotropy



(b) Distinct anomalies at
 ~ 43 K, ~ 38 K and ~ 24 K
 $T_N(\text{Mn})$ T_C $T_N'(\text{Mn})$

Dielectric anomalies at the
 magnetic transition T
profound coupling between P & M

(c) • no clear dielectric signatures
 along the a- and c-directions



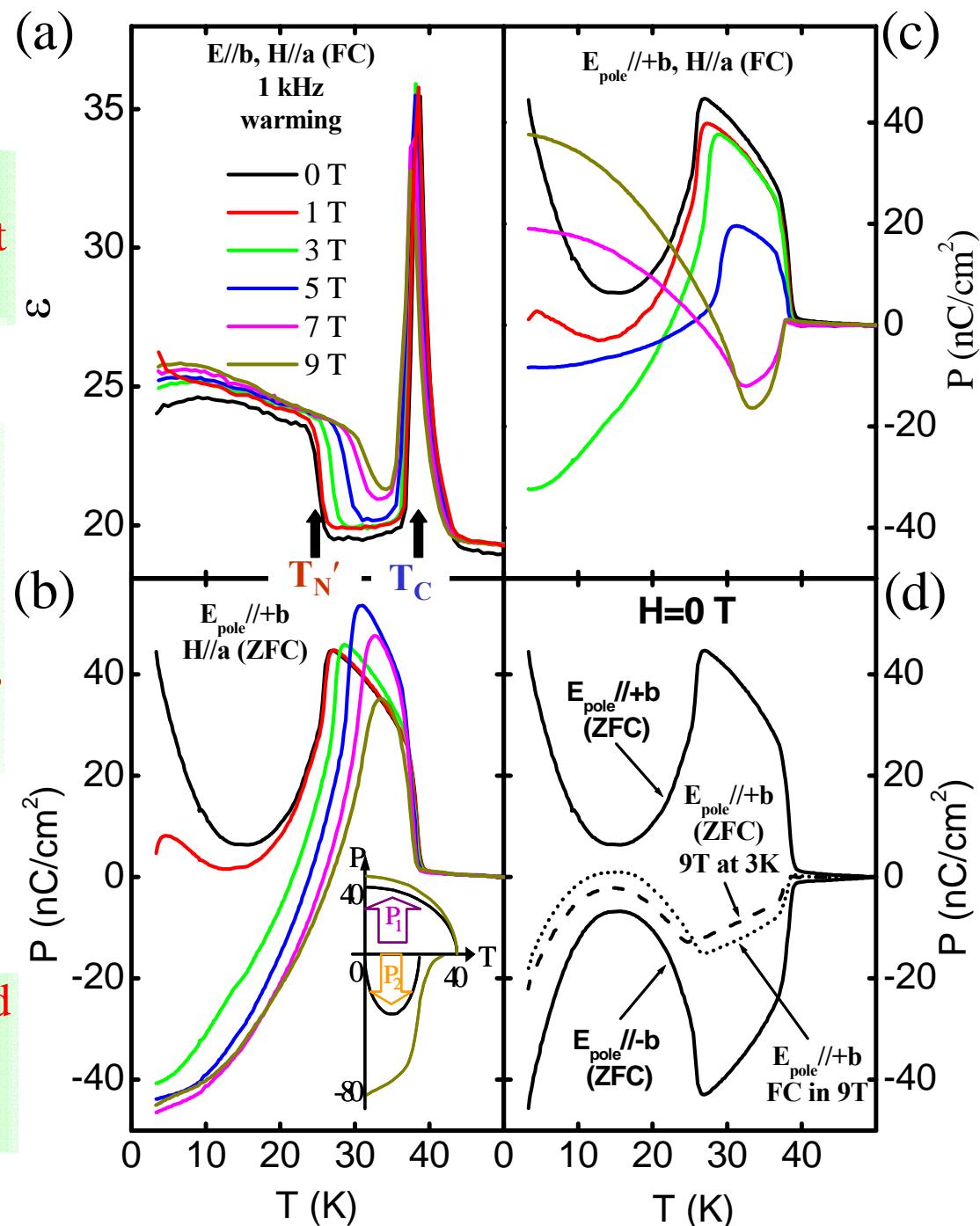
H dependence of ϵ and P

- (a) • step anomaly increases in T
• large magneto-dielectric effect near and below $T_{N'}$

- (b) • P start to increase at $T_C = 38$ K
• sudden decrease at $T_{N'}$
• complicated T dependence
→ possible model: ferrielectric two anti-parallel P components
• P reversal for $H > 2$ T

- (c) • Effect of H field cooling (FC)
→ reversed P(T) for $H \approx 7$ T

- (d) • Permanent imprint of reversed P by H even after removing H
→ “memory effect”

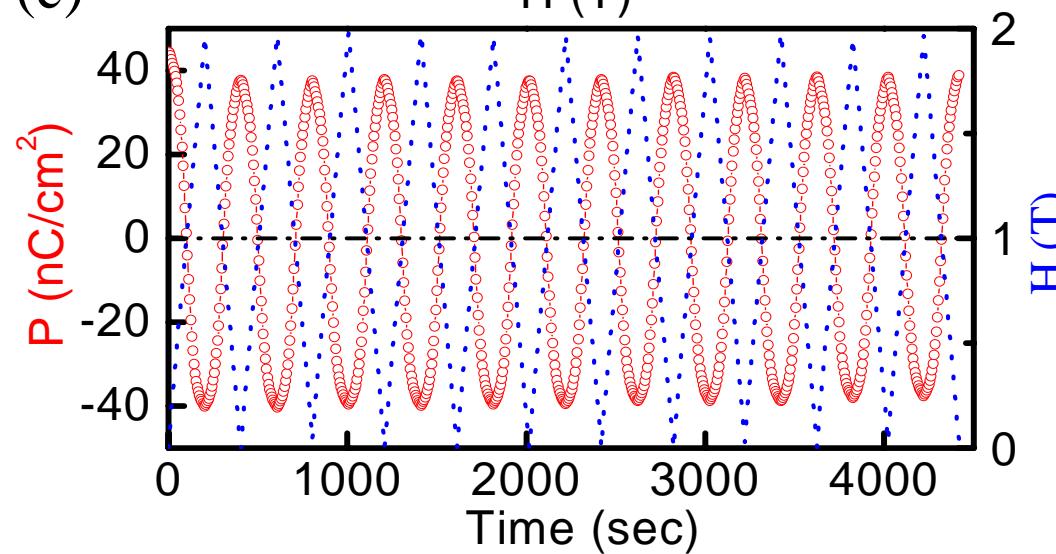
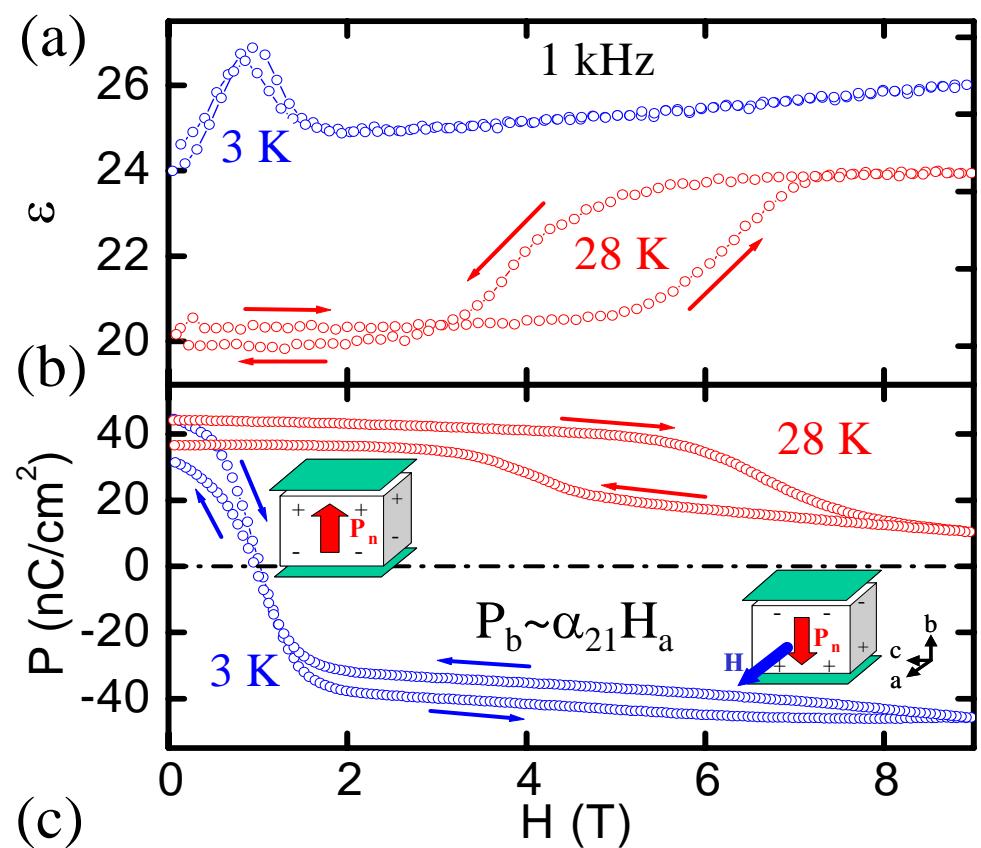


Magneto-dielectric effect and Polarization reversal

(a) large magneto-dielectric effect
13% at 3 K, 20% at 28 K

(b) • Polarization reversal by
a magnetic field of ~ 2 T
• H-induced phase transition

(c) • Sequential switching of polarization by slowly varying H from 0 to 2 T.
• **Highly reversible and reproducible** switching without any noticeable decay

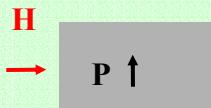


Reversible Switching by H

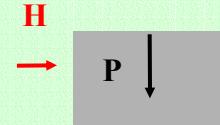
One P component ?



when $H=0$

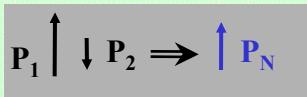


possible situation when $H \neq 0$

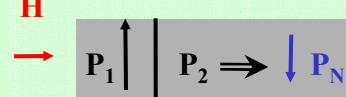


Reversible switching not possible

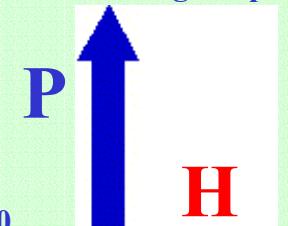
Two antiparallel P components 



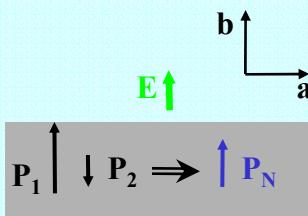
when $H=0$



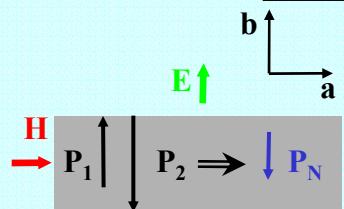
possible situation when $H \neq 0$



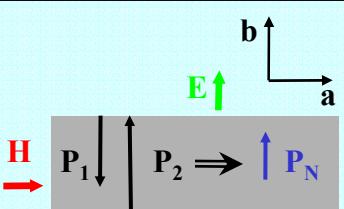
Permanent Switching by E & H



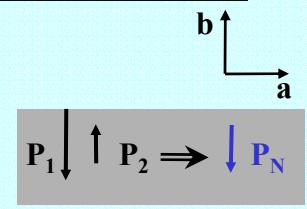
When $H=0$
P₁ is dominant



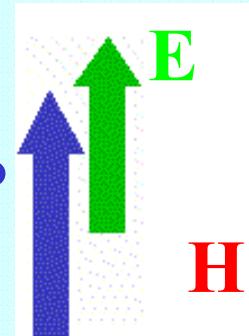
When $H \neq 0$
P₂ becomes dominant



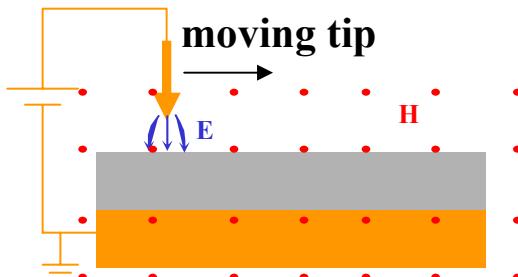
E aligns P_N along +b



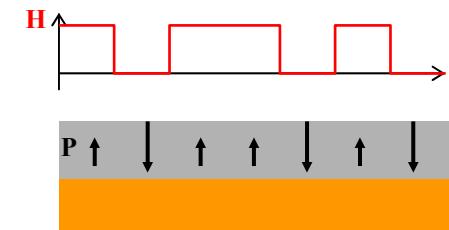
After removing E and then H
P₁ becomes dominant again
Reversed P_N imprinted



**Device application
magnetically-recordable
ferroelectric memory**



Recording of P in time varying H



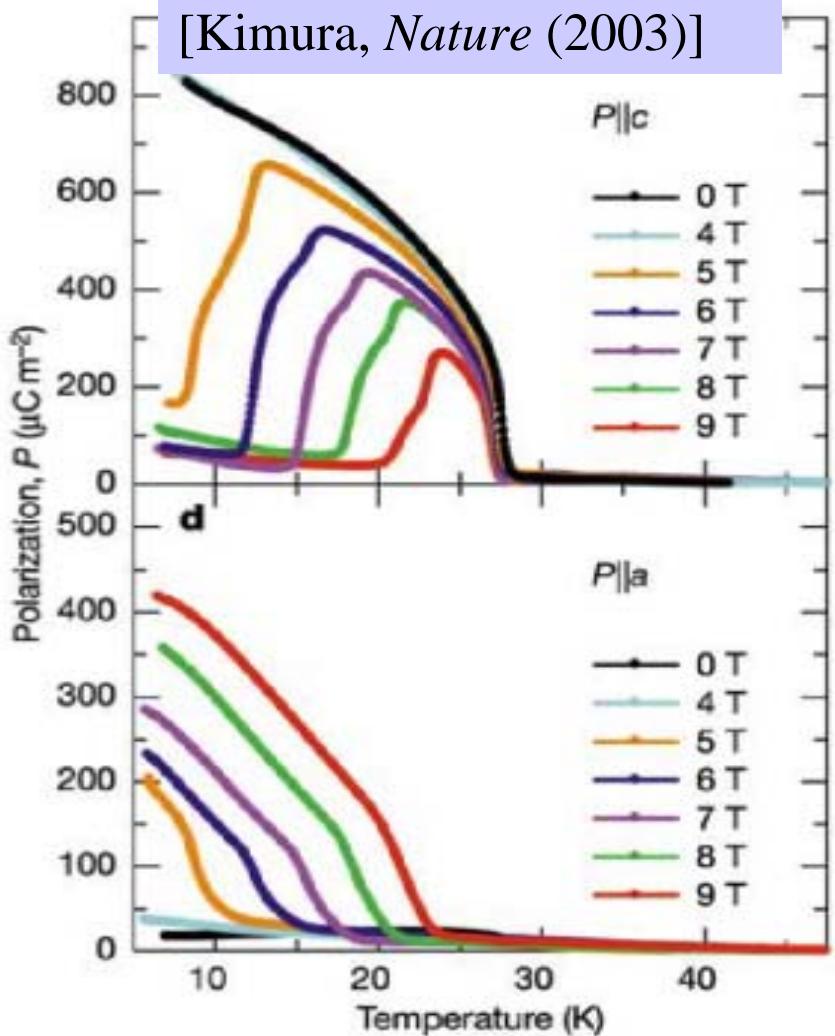
Resultant recorded P
after removing E and H

Different examples of Magnetic control of electric Polarization

TbMnO₃

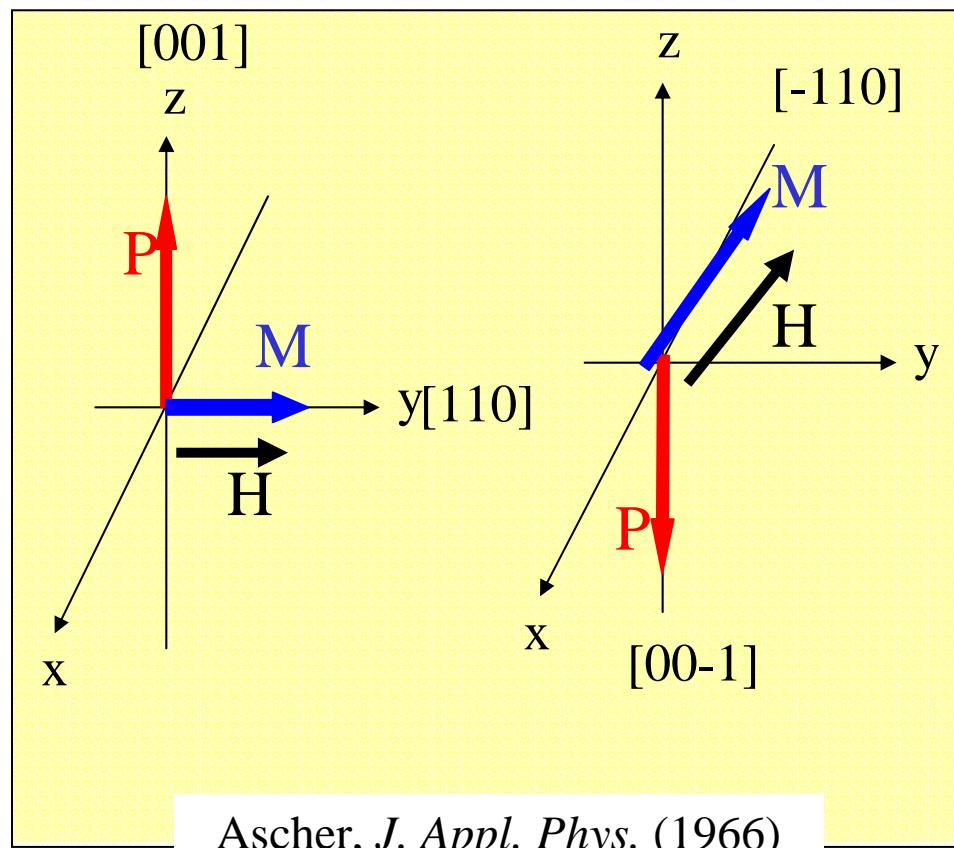
90° rotation of P by
applied $H \approx 5$ T

[Kimura, *Nature* (2003)]



Ni₃B₇O₁₃I

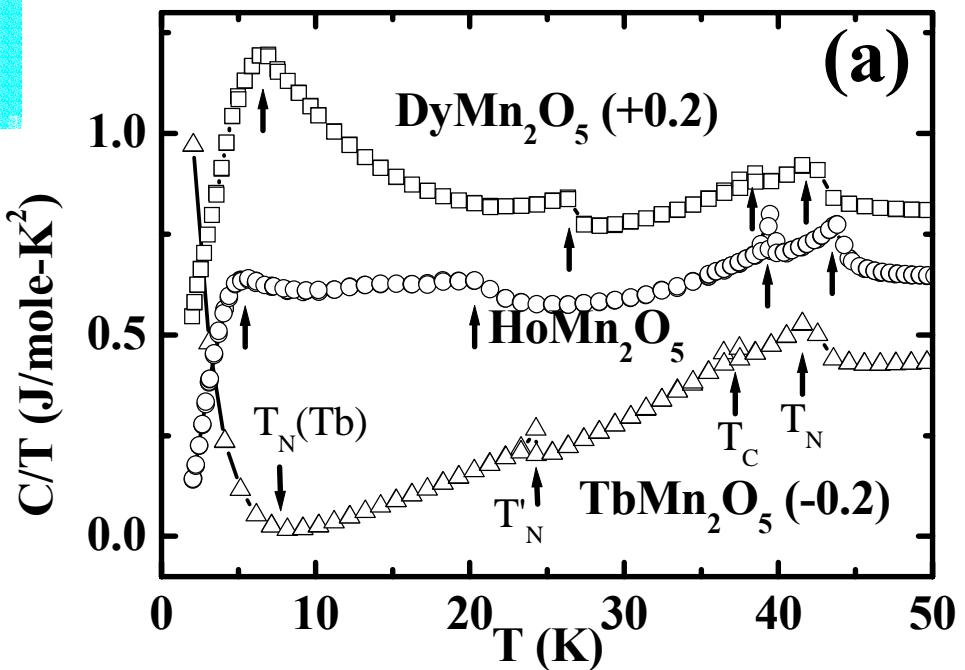
non-reversible 180° switching
by the 90° rotation of H



Ascher, *J. Appl. Phys.* (1966)

III. Colossal Magneto-Dielectric Effect in DyMn_2O_5

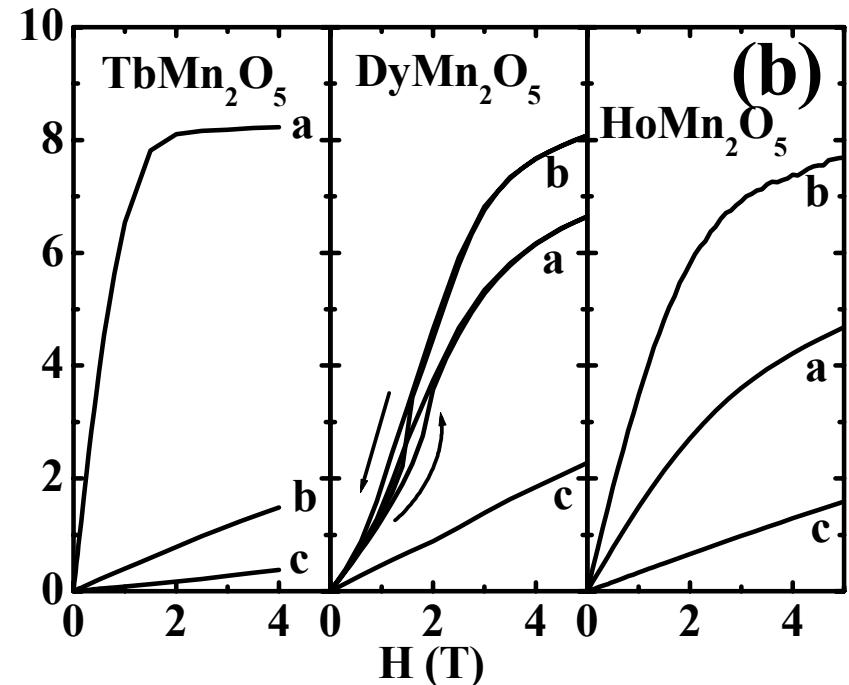
- (a) Similar set of consecutive phase transitions in REMn_2O_5
- Mn AFM $T_N \sim 40$ K
 - FE $T_C \sim 35-40$ K
 - Mn spin reorientation $T_N' \sim 20-25$ K
 - RE AFM ordering $T_N(\text{RE}) \bullet 10$ K.



- (b)
- Magnetic anisotropy
 - Systematic variation of magnetic easy direction with the size of RE ion in REMn_2O_5

Smaller RE

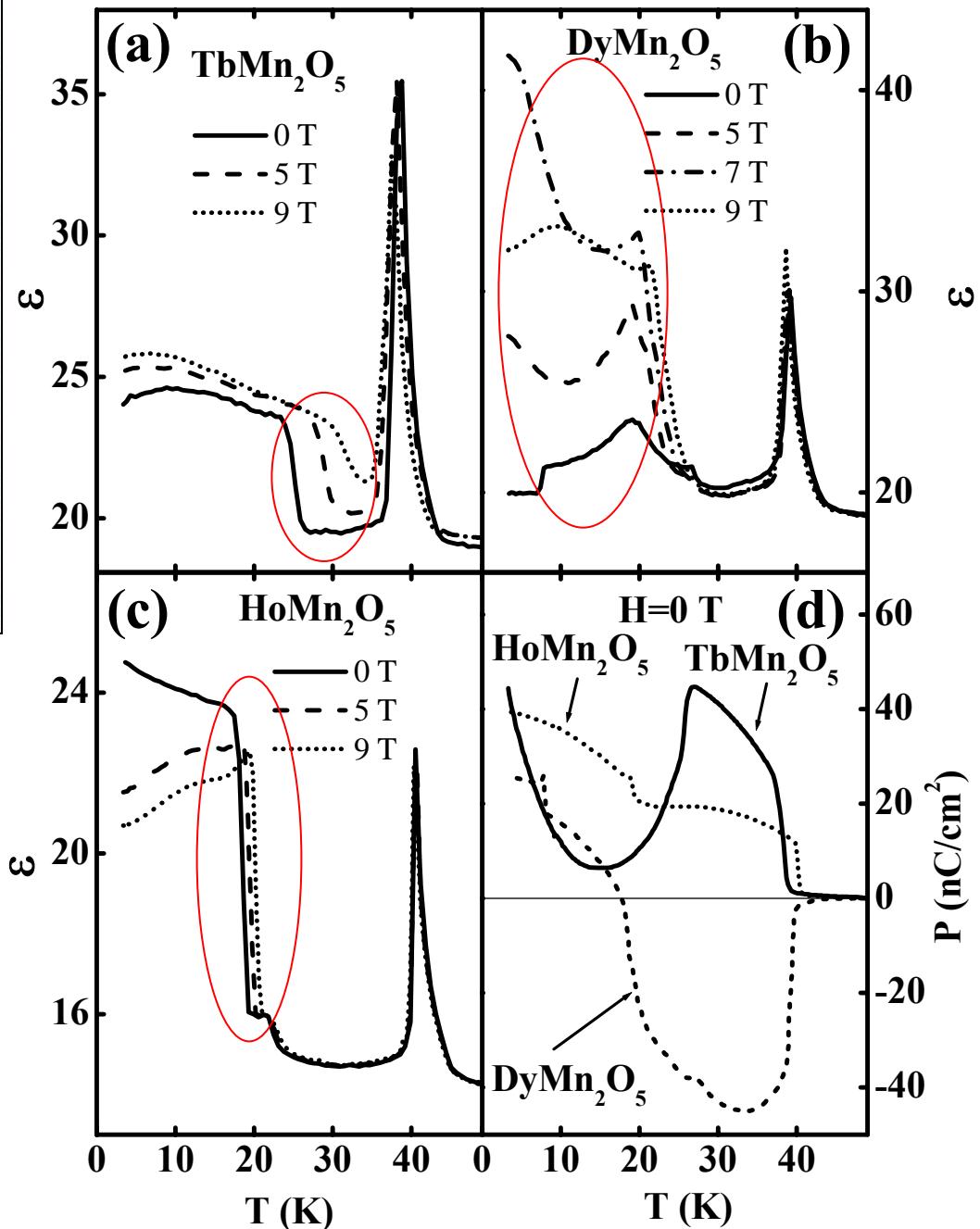
TbMn ₂ O ₅	→ <i>a</i> axis
(Dy,Ho)Mn ₂ O ₅	→ <i>b</i> axis
(Er,Tm)Mn ₂ O ₅	→ <i>c</i> axis



(a-c) Magneto-dielectric effect in REMn_2O_5

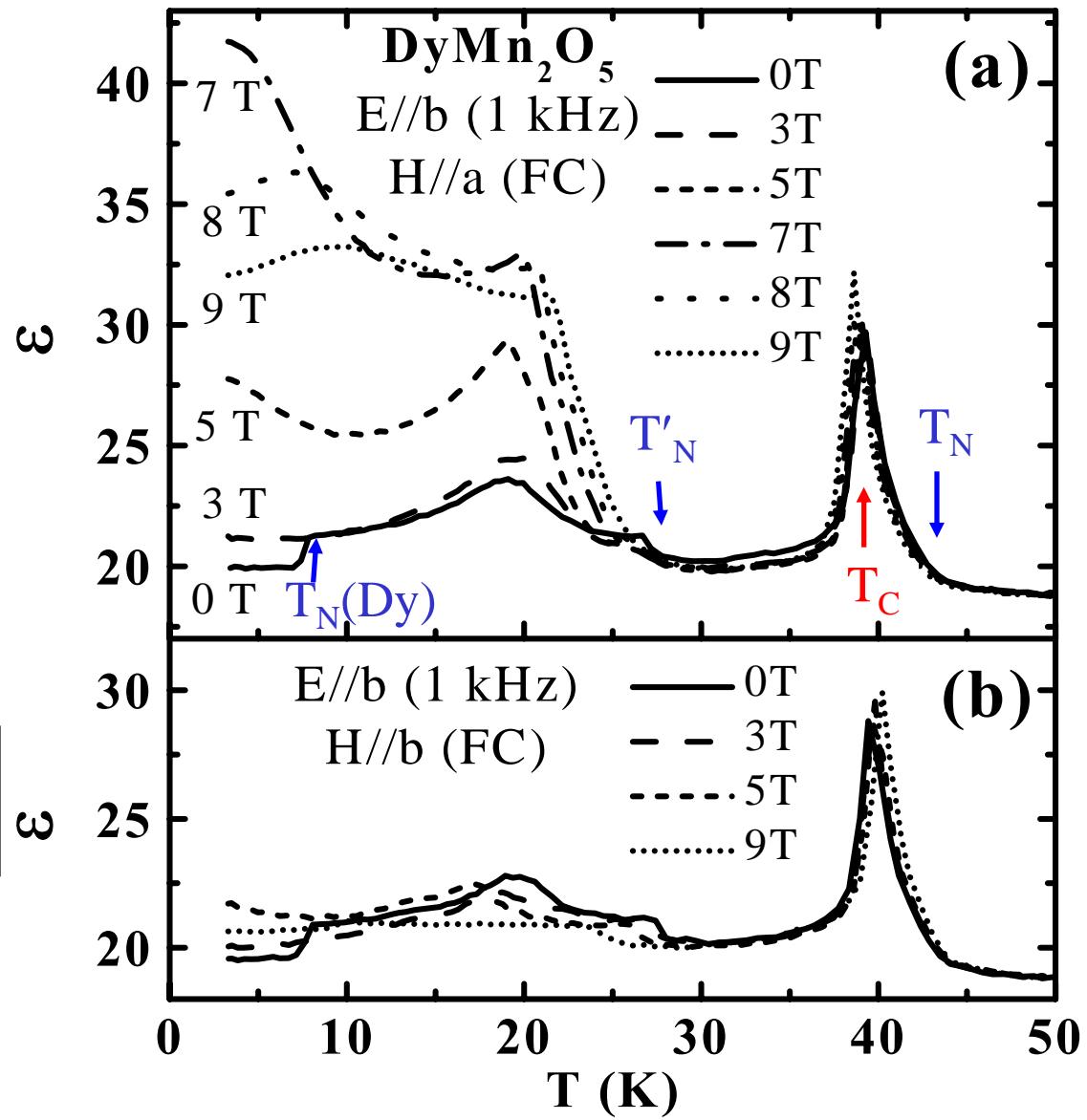
- large MD effect is observed near or below T'_N
- The largest change in ϵ 22(40) % near T'_N for $\text{Tb}(\text{Ho})\text{Mn}_2\text{O}_5$ in 9 T
- 109% at 3 K for DyMn_2O_5 in 7 T
- T'_N increases with increasing H

- (d)
- Ferrielectric model generally applicable in REMn_2O_5
 - multi-components coexist below T'_N



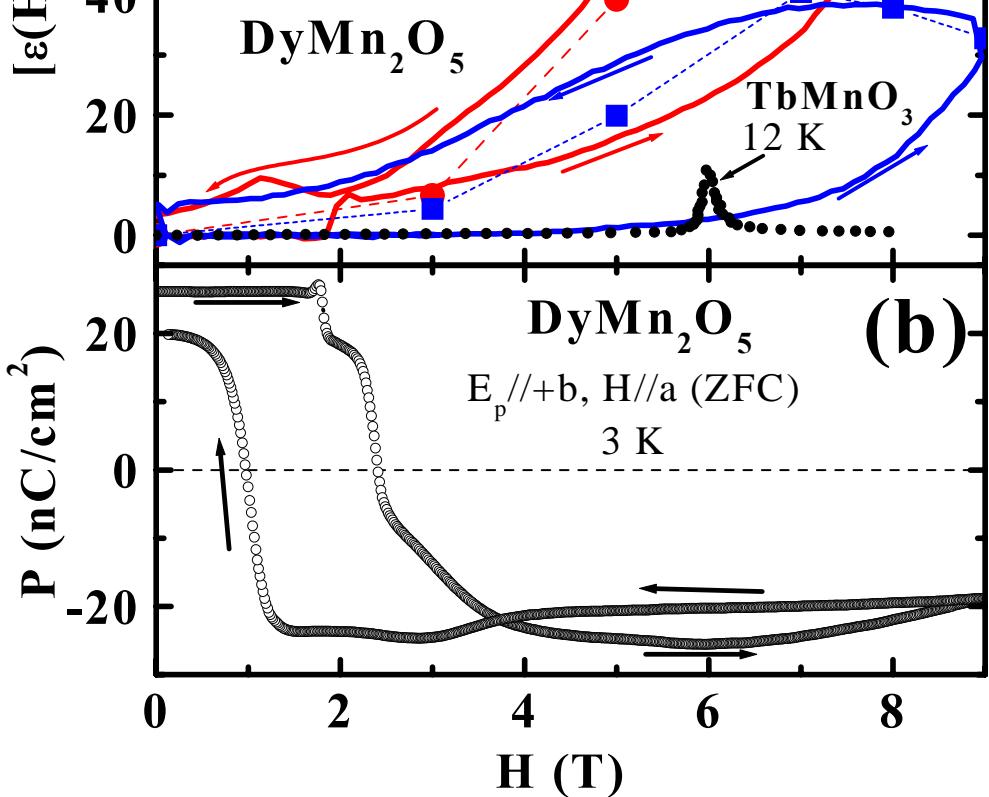
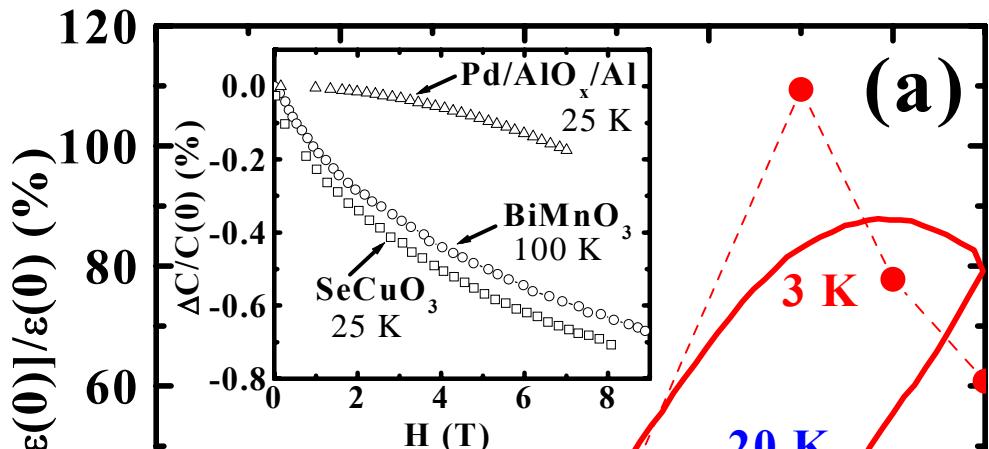
- (a) Colossal change in the dielectric constant by H
- maximum change of $\sim 109\%$ at 3 K
 - dielectric anomalies at each magnetic transition T

(b) H along the b-axis:
smaller MD effect



(a) Colossal magneto-dielectric effect in DyMn_2O_5

- maximum $\Delta\epsilon/\epsilon_0$ at 3 K
fixed field (FC): **109%**
fixed temperature (ZFC): $\sim 90\%$
- $\Delta\epsilon/\epsilon_0$ is reduced to $\sim 40\%$ at 20 K



(b) Magnetoelectric effect in DyMn_2O_5

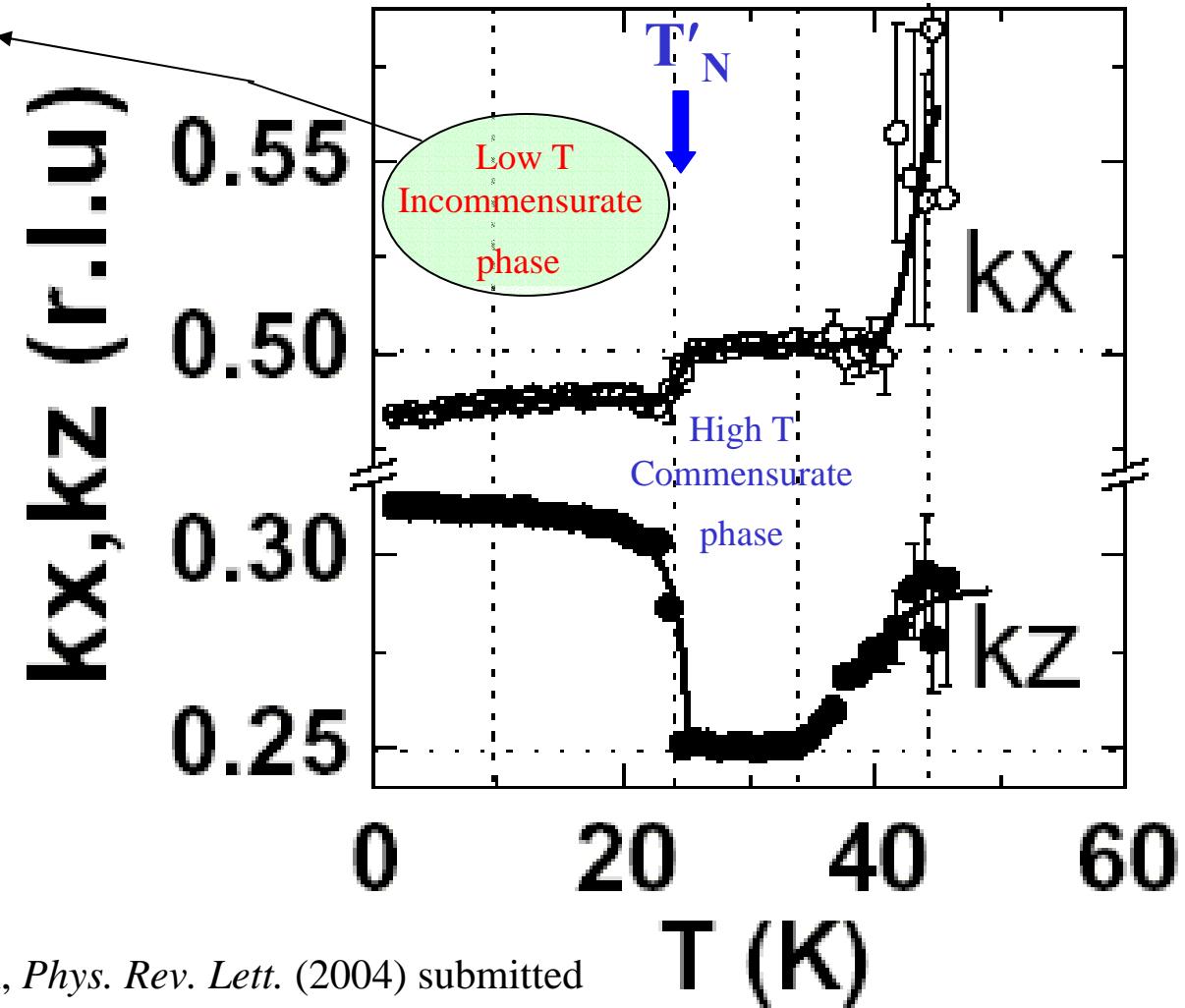
- abrupt change at ~ 1.8 T

Possible origin of colossal magneto-dielectric effect

Mn spin modulation in TbMn_2O_5
 $(k_x = 1/2 - \delta, k_y = 0, k_z = 1/4 + \gamma)$ ($\delta \approx 0.02, \gamma \approx 0.07$)

multiple P components
enormously large MD effect

Incommensurate phase
magnetically/structurally
“soft”; sensitive to external
perturbation



Summary

1. Multiferroics

Coexistence of (A)FE and (A)FM : various potential applications
Coupling between order parameters

2. Polarization reversal and memory effect by magnetic field due to the astonishing interplay between ferroelectricity and magnetism in TbMn_2O_5
3. Colossal magneto-dielectric effect in DyMn_2O_5 which appears to originate from the high sensitivity of the incommensurate state to external perturbation